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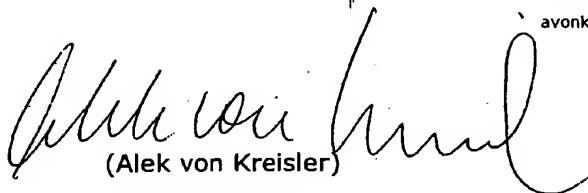
European Patent Application 03715725.2-2115
Asahi Kasei Chemicals Corporation

Enclosed is the English translation of Japanese application
no. 2002-100125 from which priority is claimed.

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I, Masanori KOMATSU, a national of Japan,
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2-1, Ohtemachi-2-chome, Chiyoda-ku, Tokyo, Japan, declare that
to the best of my knowledge and belief the attached is a full,
true, and faithful translation into English made by me of
Japanese Patent Application No. 2002-100125.

Signed this 9th day of February, 2005.


Masanori KOMATSU

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[Title of the Invention] RESIN COMPOSITION AND
MOLDED BODY

[Claims]

- 5 [Claim 1] A resin composition which contains (A) 50-
90 parts by weight of a polyamide having a relative
viscosity of not more than 3.0 measured in accordance
with JIS K6810, (B) 50-10 parts by weight of a
polyphenylene ether having a reduced viscosity of not
10 more than 0.50 and (C) a hydrogenated block copolymer
having a number-average molecular weight of not less
than 170,000 and comprising at least one polymer block
mainly composed of an aromatic vinyl compound and at
least one polymer block mainly composed of a conjugated
15 diene compound in an amount of 1-20 parts by weight
based on 100 parts by weight of the total of (A) and
(B), the proportion of the component (C) based on the
total amount of the component (B) and the component (C)
being not less than 0.10 and less than 0.30.
- 20 [Claim 2] A resin composition according to claim 1,
wherein the component (C) is a hydrogenated block
copolymer comprising at least two polymer blocks mainly
composed of an aromatic vinyl compound and at least one
polymer block mainly composed of a conjugated diene
25 compound.

[Claim 3] A resin composition according to claim 1

or 2, wherein the number-average molecular weight of one polymer block mainly composed of an aromatic vinyl compound of the component (C) is not less than 30,000.

[Claim 4] A resin composition according to any one
5 of claims 1-3, wherein the component (C) is a hydrogenated block copolymer to which an oil mainly composed of paraffin is previously added.

[Claim 5] A resin composition according to claim 1
10 which contains a transition metal as a component (D1) in an amount of not less than 10 ppm and less than 200 ppm and a halogen as a component (D2) in an amount of not less than 500 ppm and less than 1500 ppm based on the total amount of the components (A) - (C).

[Claim 6] A resin composition according to claim 1,
15 wherein the content of the component (D1) is not less than 10 ppm and less than 100 ppm and that of the component (D2) is not less than 700 ppm and less than 1200 ppm.

[Claim 7] A resin composition according to claim 1,
20 wherein the terminal amino group concentration of the polyamide of the component (A) is not more than 35 milliequivalent/kg.

[Claim 8] A resin composition according to claim 1,
wherein the component (A) is a polyamide mixture
25 containing polyamide 6 and the amount ratio of the polyamide 6 to all polyamides used is less than 15% by weight.

[Claim 9] A resin composition according to any one

of claims 1-8, wherein the component (D1) and the component (D2) are added as a master pellet of polyamide 6 base containing the (D1) component in an amount of not less than 500 ppm and the component (D2) in an amount of
5 not less than 15,000 ppm.

[Claim 10] A resin composition according to any one of claims 1-9, wherein the molecular weight distribution (weight-average molecular weight/number-average molecular weight) of polyphenylene ether of the
10 component (B) is not less than 2.5.

[Claim 11] A resin composition according to any one of claims 1-10, wherein the polyphenylene ether of the component (B) is a mixture of a polyphenylene ether having a molecular weight distribution (weight-average
15 molecular weight/number-average molecular weight) of not less than 2.5 and a polyphenylene ether having a molecular weight distribution (weight-average molecular weight/number-average molecular weight) of less than 2.5.

20 [Claim 12] An injection molded body comprising the resin composition according to any one of claims 1-11.

[Claim 13] A relay block material for automobiles which is molded from the resin composition according to any one of claims 1-12.

25 [Detailed Description of the Invention]

[0001]

[Technical Field Pertinent to the Invention]

The present invention relates to a thermoplastic resin composition which is excellent in flowability in a mold (spiral flow characteristics), less in post-deformation of molded products caused by heating, greatly inhibited from reduction of tensile strength at the time of heat aging, and further inhibited from occurrence of wrinkle pattern.

The thermoplastic resin composition of the present invention can be suitably used in a wide variety of the fields such as electric and electronic parts, OA parts, mechanical parts and electric parts, exterior trim parts and interior trim parts of motorbikes and automobiles, and can be particularly suitably used for relay block materials provided in engine rooms of automobiles.

[0002]

[Prior Art]

Polymer alloys of polyphenylene ether and polyamide have been employed for various uses since they were proposed in JP-B-45-997, and they are employed for particularly automobiles because of their excellent mechanical characteristics, thermal characteristics and fluidity characteristics.

Hitherto, polyamide 6,6 resins have been used for relay blocks provided in an engine room of automobiles, but polyamide 6,6 resins have the problem of increase in dimensional change due to absorption of water, and recently polyamide/polyphenylene ether alloys

are gradually substituted for the polyamide 6,6 resins.

For example, JP-A-6-141443, JP-A-6-136256, and JP-A-6-184398 disclose that a composition composed of a polyamide, a polyphenylene ether, polypropylene, sodium
5 montanate, etc. gives a thermoplastic resin composition which is superior in heat resistance, solvent resistance, mechanical strengths, weld strength, moldability, reusability and low-temperature shock resistance, and is suitable for automobile relay boxes.

10 [0003]

However, recently, most of automobile parts are electronically controlled, and the role of relay blocks which assemble electrical equipments becomes very important. Nevertheless, at present, the space
15 permitted for relay blocks in a highly densified engine room becomes smaller and smaller.

Thus, the structure of relay block is much complicated and the relay block has many thin wall portions, and, as a result, it is demanded that the
20 resin has higher flowability (which is not the flowability at melting shown by melt flow rate, but the flowability at the time of cooling in a mold) and is inhibited from post-deformation of the thin wall portions in a hot environment.

25 [0004]

Furthermore, there is observed such a phenomenon that wrinkle-like patterns partially occur in relay blocks designed in such a manner that thin wall

portions and thick wall portions coexist. They cause no functional problems, but give undesirable appearance.

Thus, improvement is demanded in this respect.

Moreover, owing to the high densification of
5 the inside of the engine rooms, spaces diminish, cooling efficiency lowers and further the environmental temperature in the engine rooms increases. Therefore, relay blocks are required to have the higher heat aging resistance (inhibition of reduction in tensile strength
10 during heat aging).

These problems which have been found recently cannot be sufficiently solved by the above-mentioned conventional technologies, and development of new technologies has been desired.

15 [0005]

[Problem to be solved by the Invention]

The present invention solves simultaneously the above problems of polyamide/polyphenylene ether alloys (insufficient flowability in mold, post-
20 deformation of molded products during heating, occurrence of wrinkle patterns and reduction of tensile strength during heat aging).

[0006]

[Means for Solving Problems]

25 As a result of intensive research conducted by the inventors to solve the above problems, it has been found that thermoplastic resin compositions solved in the above problems can be obtained when the composition

contains a polyamide having a specific viscosity, a polyphenylene ether having a specific viscosity and a block copolymer, in which the amount of the block copolymer is limited.

5 That is, the present invention relates to a resin composition which contains (A) 50-90 parts by weight of a polyamide having a relative viscosity of not more than 3.0 measured in accordance with JIS K6810, (B) 50-10 parts by weight of a polyphenylene ether having a
10 reduced viscosity of not more than 0.50 and (C) a hydrogenated block copolymer having a number-average molecular weight of not less than 170,000 and comprising at least one polymer block mainly composed of an aromatic vinyl compound and at least one polymer block
15 mainly composed of a conjugated diene compound in an amount of 1-20 parts by weight based on 100 parts by weight of the total of (A) and (B) , the proportion of the component (C) based on the total amount of the component (B) and the component (C) being not less than
20 0.10 and less than 0.30, and a relay block molded body comprising the above resin composition.

[0007]

Next, components usable in the present invention will be explained in detail below.

25 As polyamides of the component (A) usable in the present invention, there may be used any polyamides so long as they have an amide linkage $\{-\text{NH}-\text{C}(=\text{O})-\}$ in the polymer main chain.

In general, polyamides are obtained by ring opening polymerization of lactams, polycondensation of diamine and dicarboxylic acid, polycondensation of aminocarboxylic acids, and the like, and the polyamides
5 are not limited to those obtained in this way.

[0008]

The diamines used are roughly classified into aliphatic, alicyclic and aromatic diamines. Examples of the diamines are tetramethylenediamine,
10 hexamethylenediamine, undecamethylenediamine, dodecamethylenediamine, tridecamethylenediamine, 2,2,4-trimethylhexamethylenediamine, 2,4,4-trimethylhexamethylenediamine, 5-methylnanomethylenediamine, 1,3-bisaminomethylcyclohexane, 1,4-bisaminomethylcyclo-
15 hexane, m-phenylenediamine, p-phenylenediamine, m-xylylenediamine, p-xylylenediamine, etc.

[0009]

The dicarboxylic acids used are roughly classified into aliphatic, alicyclic and aromatic
20 dicarboxylic acids. Examples of the dicarboxylic acids are adipic acid, suberic acid, azelaic acid, sebacic acid, dodecanedioic acid, 1,1,3-tridecanedioic acid, 1,3-cyclohexanedicarboxylic acid, terephthalic acid, isophthalic acid, naphthalenedicarboxylic acid, dimer
25 acids, etc.

Examples of the lactams used are ϵ -caprolactam, enantholactam, ω -lauro lactam, etc.

Examples of the aminocarboxylic acids are ϵ -

aminocaproic acid, 7-aminoheptanoic acid, 8-amino-octanoic acid, 9-aminonanoic acid, 11-aminoundecanoic acid, 12-aminododecanoic acid, 13-aminotridecanoic acid, etc.

5 In the present invention, there may be used any copolymer polyamide obtained by polycondensation of one or two or more of the above lactams, diamines, dicarboxylic acids and ω -aminocarboxylic acids.

 Furthermore, there may be suitably used
10 polyamides obtained by polymerizing these lactams, diamines, dicarboxylic acids and ω -aminocarboxylic acids up to oligomers of low molecular weight in a polymerization reactor and then allowing them to have a high molecular weight by an extruder or the like.

15 [0010]

 As the polyamide resins (A) effectively usable in the present invention, mention may be made of polyamide 6, polyamide 6,6, polyamide 4,6, polyamide 11, polyamide 12, polyamide 6,10, polyamide 6,12, polyamide
20 6/6,6, polyamide 6/6,12, polyamide MXD (m-xylylenediamine), polyamide 6,T, polyamide 6,I, polyamide 6/6,T, polyamide 6/6,I, polyamide 6,6/6,T, polyamide 6,6/6,I, polyamide 6/6,T/6,I, polyamide 6,6/6,T/6,I, polyamide 6/12/6,T, polyamide 6,6/12/6,T,
25 polyamide 6/12/6,I, polyamide 6,6/12/6,I, etc.

 Furthermore, polyamides obtained by copolymerizing two or more polyamides by an extruder or the like can also be used. Preferred are polyamide 6, polyamide 6,6,

polyamide 6/6,6 and mixtures thereof. Polyamide 6,6 or a mixture of polyamide 6,6 and polyamide 6 is most preferred.

The amount of polyamide 6 in the mixture of polyamide 6,6 and polyamide 6 is desirably less than 15% by weight and more preferably less than 10% by weight based on all polyamides used. By using a small amount of polyamide 6, wrinkles formed on the molded piece can be markedly decreased.

10 [0011]

It is essential that the relative viscosity of the polyamide used in the present invention measured in accordance with JIS K6810 is not more than 3.0. If the viscosity of the polyamide exceeds 3.0, flowability of the composition in the mold considerably deteriorates, resulting in increase of post-deformation of the molded product after heating to cause formation of wrinkles on the molded piece. The relative viscosity of the polyamide is more preferably 2.8 or less, most preferably 2.7 or less. There is no particular lower limit, but the viscosity is desirably not less than 2.2.

[0012]

The polyamide in the present invention may be a mixture of a plurality of polyamide resins differing in relative viscosity. For example, the polyamide may be a mixture of a polyamide having a relative viscosity of 2.2 and a polyamide having a relative viscosity of 2.8, a mixture of a polyamide having a relative

viscosity of 2.7 and a polyamide having a relative viscosity of 3.2, etc. In the case of using a polyamide having a relative viscosity exceeding 3.0, the relative viscosity of the polyamide of the mixture must also be
5 3.0 or less. The relative viscosity of the polyamide mixture can be easily confirmed by actual measurement in accordance with JIS K6810.

[0013]

The terminal group of the polyamides
10 participates in the reaction with polyphenylene ether made functional. Polyamides generally have an amino group or carboxyl group as a terminal group, and, in general, impact resistance decreases and flowability improves with increase of the concentration of carboxyl
15 group and, on the other hand, impact resistance increases and flowability deteriorates with increase of the concentration of amino group.

In the present invention, the ratio of these terminal groups, namely, amino group/carboxyl group in
20 concentration is preferably $9/1-1/9$, more preferably $8/2-1/9$, further preferably $6/4-1/9$.

[0014]

The concentration of the terminal amino group is preferably not more than 50 milliequivalent/kg, more
25 preferably not more than 40 milliequivalent/kg, most preferably not more than 35 milliequivalent/kg. There is no particular limitation in the lower limit, but it is desirably 10 milliequivalent/kg or more. When the

concentration of the terminal amino group exceeds 50 milliequivalent/kg, considerable deterioration of flowability of the composition in a mold, increase of post-deformation of the molded product after heating and
5 occurrence of a wrinkle-pattern on the molded piece are apt to occur.

These terminal groups of polyamide resins can be adjusted by a method known to one skilled in the art. For example, at least one compound selected from diamine
10 compounds, monoamine compounds, dicarboxylic acid compounds, monocarboxylic acid compounds, etc. is added at the time of polymerization of the polyamide resin so as to give a desired terminal group concentration.

Moreover, in the present invention, for the
15 purpose of further improvement of heat stability imparted by the polyamide resin to the resin composition, a transition metal and a halogen may be contained in the resin composition. The transition metal is not particularly limited, but copper, cerium,
20 nickel and cobalt are preferred, and copper is especially preferred. As halogens, bromine or iodine is preferred.

[0015]

The amount of transition metal is preferably
25 not less than 10 ppm and less than 200 ppm, more preferably not less than 10 ppm and less than 100 ppm in the resin composition. The amount of halogen is preferably not less than 500 ppm and less than 1500 ppm,

more preferably not less than 700 ppm and less than 1200 ppm.

The method for adding the transition metal and halogen to the resin composition includes, for example,
5 a method of adding them in the form of a powder at the time of melt kneading of the composition of polyamide/polyphenylene ether, a method of adding them at the time of polymerization of the polyamide, a method of preparing a master pellet by adding them to the
10 polyamide at a high concentration and then adding the master pellet to the resin composition, and other methods, and any of these methods can be employed. Of these methods, preferred are the method of adding them at the time of polymerization of the polyamide and the
15 method of preparing a master pellet by adding them to the polyamide at a high concentration and then adding the master pellet to the resin composition, and the latter method is most preferred.

[0016]

20 Among the methods of preparing a master pellet by adding them to a polyamide at a high concentration and then adding the master pellet to the resin composition, most preferred is a method of addition in the form of a master pellet of polyamide 6 as a base
25 which contains 500 ppm or more of transition metal and 15,000 ppm or more of halogen. Use of this method can further inhibit deterioration of tensile strength characteristics after heat aging. The reason is not

clear, but one factor is considered that when a transition metal or halogen is present in polyamide 6 of low crystallinity, ionization readily occurs, which is apt to contribute to stabilization of the polyamide.

5 [0017]

Moreover, in the present invention, in addition to the above materials, known organic stabilizers can also be used without any problems. Examples of the organic stabilizers are hindered
10 phenolic antioxidants such as Irganox 1098, phosphorus-based processing heat stabilizers such as Irgafos 168, lactone-based processing heat stabilizers such as HP-136, sulfur-based heat stabilizers, hindered amine-based light stabilizers, etc.

15 Of these organic stabilizers, hindered phenolic antioxidants, phosphorus-based processing heat stabilizers or combinations thereof are more preferred.

The amount of organic stabilizers is preferably 0.001-1 part by weight based on 100 parts by
20 weight of polyamide resin.

[0018]

In addition to the above additives, other known additives which can be added to polyamides may be added in an amount of less than 10 parts by weight based
25 on 100 parts by weight of polyamide.

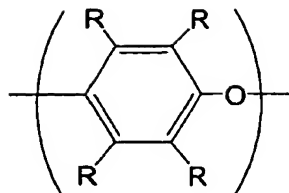
The polyphenylene ether usable in the present invention is a homopolymer and/or copolymer comprising the following structural unit of the following formula

(1):

[0019]

[Formula 1]

5



(1)

[0020]

10 [in the above formula, O represents an oxygen atom and R independently of one another represents hydrogen, halogen, primary or secondary lower alkyl, phenyl, haloalkyl, aminoalkyl, hydrocarbonoxy or
15 carbon atoms separate the halogen atom and oxygen atom)].

[0021]

Specific examples of the polyphenylene ether (B) of the present invention are poly(2,6-dimethyl-1,4-
20 phenylene ether), poly(2-methyl-6-ethyl-1,4-phenylene ether), poly(2-methyl-6-phenyl-1,4-phenylene ether), poly(2,6-dichloro-1,4-phenylene ether), etc., and further include polyphenylene ether copolymers such as copolymers of 2,6-dimethylphenol with other phenols (for
25 example, copolymers of 2,6-dimethylphenol with 2,3,6-trimethylphenol or 2-methyl-6-butylphenol as disclosed in JP-B-52-17880).

[0022]

Among them, especially preferred are poly(2,6-dimethyl-1,4-phenylene ether), copolymers of 2,6-dimethylphenol with 2,3,6-trimethylphenol or mixtures thereof.

5 The method for producing the polyphenylene ether (B) used in the present invention is not particularly limited and includes known methods. For example, there may be used those which are disclosed in U.S. Patent Nos. 3306874, 3306875, 3257357 and 3257358,
10 JP-A-50-51197, JP-B-52-17880 and JP-B-63-152628.

[0023]

The reduced viscosity of polyphenylene ether in the present invention η_{sp}/c : measured using a 0.5 g/dl chloroform solution at 30°C) is essentially not more
15 than 0.5 dl/g, preferably not more than 0.48 dl/g, more preferably not more than 0.45 dl/g. There is no lower limit, but not less than 0.30 dl/g is preferred.

In the present invention, a mixture of two or more polyphenylene ethers differing in reduced viscosity
20 can be used suitably so long as the reduced viscosity of the mixture is not more than 0.5 dl/g. The reduced viscosity of mixture can be easily measured by measuring it as a chloroform solution of 0.5 dl/g at 30°C.

[0024]

25 The polyphenylene ether used in the present preferably has a molecular weight distribution (weight-average molecular weight/number-average molecular weight) of not less than 2.5. As the upper limit of the

molecular weight distribution, it is preferably less than 3.5. By using a polyphenylene ether having a molecular weight distribution of not less than 2.5, the flowability in the mold can further be improved.

5 Furthermore, it is preferred to use a mixture of a polyphenylene ether having a molecular weight distribution (weight-average molecular weight/number-average molecular weight) of not less than 2.5 and less than 3.5 and a polyphenylene ether having a molecular
10 weight distribution of less than 2.5 and not less than 1.5. By using the mixture, not only the flowability in the mold is improved, but also occurrence of the wrinkle patterns can be inhibited.

[0025]

15 The molecular weight distribution in the present invention is expressed by a ratio of weight-average molecular weight and number-average molecular weight (weight-average molecular weight/number-average molecular weight) measured by an ultraviolet
20 spectrodetector using a gel permeation chromatography measuring apparatus and calculated in terms of standard polystyrene.

Furthermore, in the polyphenylene ether usable in the present invention, an organic solvent used as a
25 polymerization solvent may remain in an amount of less than 5% by weight based on 100 parts by weight of polyphenylene ether. The organic solvent remaining after it is used as a polymerization solvent is

difficult to completely remove at the drying step after polymerization, and usually remains in an amount of from several hundred ppm to several percent. Examples of the organic solvent used as a solvent for polymerization of polyphenylene ether are at least one member from isomers of toluene or xylene, ethylbenzene, alcohols of 1-5 carbon atoms, chloroform, dichloromethane, chlorobenzene, dichlorobenzene, etc.

[0026]

Moreover, the polyphenylene ether usable in the present invention may be a wholly or partly modified polyphenylene ether.

The modified polyphenylene ether here means a polyphenylene ether modified with at least one modifying compound which has at least one carbon-carbon double bond or triple bond and at least one carboxylic acid group, acid anhydride group, amino group, hydroxyl group or glycidyl group in the molecular structure.

[0027]

For producing the modified polyphenylene ether, in the presence or absence of a radical initiator, (1) polyphenylene ether is reacted with a modifying compound at a temperature of 100°C or higher and lower than the glass transition temperature of the polyphenylene ether without melting the polyphenylene ether, (2) polyphenylene ether is melt kneaded and reacted with a modifying compound at a temperature of not lower than the glass transition temperature of the

polyphenylene ether and not higher than 360°C, or (3)
polyphenylene ether is reacted with a modifying compound
in a solution at a temperature lower than the glass
transition temperature of the polyphenylene ether. Any
5 of these methods can be used, but the method (1) or (2)
is preferred.

[0028]

Next, at least one modifying compound which
has at least one carbon-carbon double bond or triple
10 bond and at least one carboxylic acid group, acid
anhydride group, amino group, hydroxyl group or glycidyl
group in the molecular structure will be specifically
explained.

Examples of modifying compounds having
15 simultaneously carbon-carbon double bond and carboxylic
acid group or acid anhydride group in the molecule are
maleic acid, fumaric acid, chloromaleic acid, cis-4-
cyclohexene-1,2-dicarboxylic acid and acid anhydrides
thereof. Among them, fumaric acid, maleic acid and
20 maleic anhydride are preferred, and fumaric acid and
maleic anhydride are especially preferred.

[0029]

Moreover, one or two of the two carboxyl
groups of these unsaturated dicarboxylic acids may be
25 esterified.

Examples of modifying compounds having
simultaneously carbon-carbon double bond and glycidyl
group in the molecule are allyl glycidyl ether, glycidyl

acrylate, glycidyl methacrylate, epoxidized natural fats and oils, etc.

Among them, glycidyl acrylate and glycidyl methacrylate are especially preferred.

- 5 Examples of modifying compounds having simultaneously carbon-carbon double bond and hydroxyl group in the molecule are unsaturated alcohols of the formula $C_nH_{2n-3}OH$ (n is a positive integer) such as allyl alcohol, 4-pentene-1-ol and 1,4-pentadiene-3-ol,
- 10 unsaturated alcohols of the formulas $C_nH_{2n-5}OH$ and $C_nH_{2n-7}OH$ (n is a positive integer), and the like.

The above modifying compounds may be used each alone or in combination of two or more.

[0030]

- 15 The amount of modifying compound used for preparation of modified polyphenylene ether is preferably 0.1-10 parts by weight, more preferably 0.3-5 parts by weight based on 100 parts by weight of polyphenylene ether.

- 20 Preferred amount of radical initiator used for preparation of radical initiator-modified polyphenylene ether is 0.001-1 part by weight based on 100 parts by weight of polyphenylene ether.

[0031]

- 25 Furthermore, the addition rate of the modifying compound to the modified polyphenylene ether is preferably 0.01-5% by weight, more preferably 0.1-3% by weight.

In the modified polyphenylene ether, unreacted modified compound and/or polymer of the modifying compound may remain in an amount of less than 1% by weight.

5 In order to reduce the amount of unreacted modifying compound and/or polymer of the modifying compound remaining in the modified polyphenylene ether, if necessary, a compound having an amide linkage and/or amino group may be added in preparation of the modified
10 polyphenylene ether.

[0032]

The compound having an amide linkage is intended to mean a compound having the amide linkage $\{-\text{NH}-\text{C}(=\text{O})-\}$ structure in the molecular structure, and
15 the compound having an amino group is intended to mean a compound having the $\{-\text{NH}_2\}$ structure at the terminal. Specific examples of these compounds are aliphatic amines such as octylamine, nonylamine, tetramethylenediamine and hexamethylenediamine, aromatic
20 amines such as aniline, m-phenylenediamine, p-phenylenediamine, m-xylylenediamine and p-xylylenediamine, reaction products of these amines with carboxylic acids, dicarboxylic acids, etc., lactams such as ϵ -caprolactam, polyamide resins, and the like, but
25 the compound is not limited to these examples.

[0033]

The amount of the addition of the compound having an amide linkage and/or amino group is preferably

not less than 0.001 part by weight and less than 5 parts by weight, more preferably not less than 0.01 part by weight and less than 1 part by weight, further preferably not less than 0.01 part by weight and less than 0.1 part by weight based on 100 parts by weight of polyphenylene ether.

Moreover, in the present invention, a styrenic thermoplastic resin may be added in an amount of less than 50 parts by weight based on 100 parts by weight of polyamide and polyphenylene ether in total.

[0034]

Examples of the styrenic thermoplastic resins in the present invention are homopolystyrenes, rubber-modified polystyrenes (HIPS), styrene-acrylonitrile copolymers (AS resin), styrene - rubber-like polymer - acrylonitrile copolymers (ABS resin), etc.

Further, various stabilizers which are known for stabilization of polyphenylene ether can be suitably used. Examples of the stabilizers are metallic stabilizers such as zinc oxide and zinc sulfide, and organic stabilizers such as hindered phenolic stabilizers, phosphorus-based stabilizers and hindered amine stabilizers, and the preferred amount of the stabilizers is less than 5 parts by weight based on 100 parts by weight of polyphenylene ether.

In addition, known additives that can be added to polyphenylene ether may be added in an amount of less than 10 parts by weight based on 100 parts by weight of

polyphenylene ether.

[0035]

As to the weight ratio of polyamide and polyphenylene ether in the present invention, polyamide
5 is in an amount of 50-90 parts by weight and polyphenylene ether is in an amount of 10-50 parts by weight (total of them being 100 parts by weight). If the amount of polyphenylene ether exceeds 50 parts by weight, the flowability in the mold is deteriorated,
10 which is not preferred.

[0036]

The block copolymer (C) used in the present invention will be explained specifically.

The block copolymer (C) used in the present
15 invention is a hydrogenated block copolymer having a number-average molecular weight of not less than 170,000 and comprising at least one polymer block mainly composed of an aromatic vinyl compound and at least one polymer block mainly composed of a conjugated diene
20 compound.

Specific examples of the aromatic vinyl compound are styrene, α -methylstyrene, vinyltoluene, etc., and one or more compounds selected from them are used. Styrene is particularly preferred.

25 Specific examples of the conjugated diene compound are butadiene, isoprene, piperylene, 1,3-pentadiene, etc., and one or more compounds selected from them are used. Butadiene, isoprene or a

combination of them is preferred.

[0037]

When butadiene is used as the conjugated diene compound of the block copolymer, the microstructure of the polybutadiene block portion preferably contains 5-80% of 1,2-vinyl or 1,2-vinyl and 3,4-vinyl in total, and this is more preferably 10-50%, most preferably 15-40%.

The block copolymer comprising polymer block (a) mainly composed of an aromatic vinyl compound and polymer block (b) mainly composed of a conjugated diene compound in the present invention is preferably a block copolymer having a bonding type selected from a-b type, a-b-a type and a-b-a-b type.

[0038]

Among these types, more preferred are a-b-a type and a-b-a-b type, and most preferred is a-b-a type. These may be mixtures.

The hydrogenated block copolymer used in the present invention is intended to mean the above-mentioned block copolymer comprising polymer block of aromatic vinyl compound and polymer block of conjugated diene compound in which the amount of aliphatic double bonds is controlled to the range of more than 0% and up to 100% by hydrogenation of the block copolymer. The hydrogenation rate of the hydrogenated block copolymer is preferably 50% or higher, more preferably 80% or higher and most preferably 95% or higher.

[0039]

In the present invention, it is necessary that the block copolymer of the component (C) used in the present invention is a block copolymer having a number-average molecular weight of not less than 170,000.

When a block copolymer having a number-average molecular weight of less than 170,000 is used, the tensile strength before heat aging which is the basis for the tensile strength after heat aging as one of the effects of the present invention is deteriorated undesirably.

The number-average molecular weight defined in the present invention is intended to mean a number-average molecular weight measured by an ultraviolet spectrodetector using a gel permeation chromatography measuring apparatus and calculated in terms of standard polystyrene. In this case, there may be detected low molecular weight components due to deactivation of catalyst during polymerization, and in such a case, the low molecular weight components are not included in the calculation of the molecular weight. Typically, the calculated exact molecular weight distribution (weight-average molecular weight/number-average molecular weight) is within the range of 1.0-1.1.

[0040]

In the present invention, the number-average molecular weight of one polymer block mainly composed of aromatic vinyl compound in the component (C) is





desirably not less than 30,000. When the number-average molecular weight of one polymer block mainly composed of aromatic vinyl compound is not less than 30,000, reduction of tensile strength after heat aging can be
5 inhibited. That is because the aromatic vinyl compound block of the block copolymer has a molecular weight enough to compatibilize with polyphenylene ether, heat resistance of the block copolymer is improved to inhibit heat deterioration.

10 [0041]

The number-average molecular weight of one polymer block mainly composed of aromatic vinyl compound can be determined by the following formula using the above-mentioned number-average molecular weight of the
15 block copolymer.

$$Mn_{(a)} = \{Mn \times a / (a+b)\} / N$$

[in the above formula, $Mn_{(a)}$ denotes the number-average molecular weight of one polymer block mainly composed of aromatic vinyl compound, Mn denotes the number-average molecular weight of the block copolymer, a denotes a
20 weight percent of all polymer blocks mainly composed of aromatic vinyl compound in the block copolymer, b denotes a weight percent of all polymer blocks mainly composed of conjugated diene compound in the block copolymer, and N denotes the number of polymer blocks
25 mainly composed of aromatic vinyl compound in the block

copolymer].

[0042]

In the present invention, the amount of the block copolymer of the component (C) is 1-20 parts by weight based on 100 parts by weight of the polyamide and the polyphenylene ether in total.

Furthermore, it is necessary that the proportion of the block copolymer of the component (C) based on the total amount of the polyphenylene ether of the component (B) and the component (C) is not less than 0.10 and less than 0.30. The proportion is more preferably not less than 0.10 and less than 0.20, further preferably not less than 0.10 and less than 0.15.

If the proportion is less than 0.10, wrinkles are apt to occur on the molded piece, and if it is 0.30 or more, the tensile strength before heat aging which is the basis for the tensile strength after heat aging as one of the effects of the present invention is deteriorated undesirably.

[0043]

Without departing from the spirit of the present invention, these block copolymers comprising block of aromatic vinyl compound and block of conjugated diene compound used in the present invention may be used in combination of two or more which differ in bond form, kind of aromatic vinyl compound, kind of conjugated diene compound, amount of 1,2-vinyl bond or amount of

1,2-vinyl bond and 3,4-vinyl bond, content of aromatic vinyl compound component and hydrogenation rate.

[0044]

Furthermore, these block copolymers used in
5 the present invention may be wholly or partially modified block copolymers.

The modified block copolymer is intended to mean a block copolymer modified with at least one modifying compound which has at least one carbon-carbon
10 double bond or triple bond and at least one carboxylic acid group, acid anhydride group, amino group, hydroxyl group or glycidyl group in the molecular structure.

[0045]

For producing the modified block copolymer, in
15 the presence or absence of radical initiator, (1) the block copolymer is melt kneaded and reacted with a modifying compound at a temperature of not lower than the softening point of the block copolymer and not higher than 250°C, (2) the block copolymer and a
20 modifying compound are reacted in a solution at a temperature of not higher than the softening point of the block copolymer, or (3) the block copolymer is reacted with a modifying compound at a temperature of not higher than the softening point of the block
25 copolymer without melting the block copolymer and the modifying compound. Any of these methods can be used, but the method (1) is preferred, and the method (1) carried out in the presence of a radical initiator is

most preferred.

[0046]

As at least one modifying compound having at least one carbon-carbon double bond or triple bond and
5 at least one carboxylic acid group, acid anhydride group, amino group, hydroxyl group or glycidyl group in the molecular structure, there may be used the same modifying compounds as mentioned above in connection with modification of polyphenylene ether.

10 Furthermore, the block copolymer of the present invention may previously contain an oil mainly composed of paraffin. By previously adding an oil mainly composed of paraffin, the effects to improve flowability in the mold and inhibit occurrence of post-
15 deformation of molded products during heating and formation of wrinkle patterns can be further enhanced.

[0047]

The amount of the oil mainly composed of paraffin is less than 70 parts by weight, more
20 preferably less than 60 parts by weight based on 100 parts by weight of the block copolymer of the component (C).

The oil mainly composed of paraffin is a mixture of hydrocarbon compounds having a weight-average
25 molecular weight of 500-10000 which comprises three components of an aromatic ring-containing compound, a naphthene ring-containing compound and a paraffin compound, and has a content of the paraffin compound of

50% by weight or more.

[0048]

More preferably, the oil mainly composed of paraffin comprises a combination of 50-90% by weight of a paraffinic compound, 10-40% by weight of a naphthene ring-containing compound and 5% by weight or less of an aromatic ring-containing compound. One example of commercially available oils mainly composed of paraffin is PW-380 (manufactured by Idemitsu Kosan Co., Ltd.).

Further, a compatibilizer may be added at the time of preparing the resin composition of the present invention. A main purpose for using a compatibilizer is to improve the physical properties of the polyamide-polyphenylene ether mixture. The compatibilizer used in the present invention is intended to mean a polyfunctional compound which interacts with polyphenylene ether, polyamide or both of them. This interaction may be a chemical one (for example, grafting) or a physical one (for example, change in surface characteristics of the disperse phase).

[0049]

Either way, the resulting polyamide-polyphenylene ether mixture shows improved compatibility.

Examples of the compatibilizer usable in the present invention are mentioned in detail in JP-A-8-8869, JP-A-9-124926, etc. and all of these known compatibilizers can be used and may also be used in

combination.

Among these various compatibilizers, particularly suitable ones are, for example, maleic acid, maleic anhydride and citric acid.

5 The amount of compatibilizer in the present invention is preferably 0.01-20 parts by weight, more preferably 0.1-10 parts by weight based on 100 parts by weight of the mixture of polyamide and polyphenylene ether.

10 [0050]

In the present invention, if necessary, other additional components may be added so long as the effect of the present invention is not damaged.

Examples of the additional components are
15 other thermoplastic resins such as polyester and polyolefin, inorganic fillers (such as talc, kaolin, xonotlite, wollastonite, titanium oxide, potassium titanate, carbon fibers and glass fibers), known silane coupling agents for enhancing affinity between inorganic
20 fillers and resins, flame retardants (such as halogenated resins, silicone flame retardants, magnesium hydroxide, aluminum hydroxide, organic phosphate ester compounds, ammonium polyphosphate, melamine polyphosphate and red phosphorus), fluorine-based polymers having an
25 effect to inhibit dripping, plasticizers (such as oils, low molecular weight polyolefin, polyethylene glycol and fatty acid esters), flame retardation assistants such as antimony trioxide, coloring agents such as carbon black,

electric conductivity imparting agents such as carbon fibers, conductive carbon black and carbon fibrils, antistatic agents, various peroxides, antioxidants, ultraviolet absorbers, light stabilizers, etc.

5 [0051]

The specific amount of these components is not more than 100 parts by weight based on 100 parts by weight of the components (A)-(C) in total.

Examples of the processing machines for
10 obtaining the composition of the present invention are single-screw extruders, twin-screw extruders, rolls, kneaders, Brabender Plastograph, Banbury mixers, etc., and twin-screw extruders are preferred, and twin-screw extruders having an upstream side feed opening and one
15 or more downstream side feed openings are most preferred.

[0052]

The melt kneading temperature for obtaining the resin composition of the present invention is not
20 particularly limited, and considering the kneading state, conditions for obtaining suitable compositions can be optionally selected usually from 240-360°C.

[0053]

As the specific production method according to
25 the present invention, mention may be made of the following methods which use a twin-screw extruder having an upstream side feed opening and one or more downstream side feed openings: (1) a method which comprises feeding

a block copolymer and a polyphenylene ether from the upstream side feed opening and melt kneading them, and feeding then a polyamide from the downstream side feed opening, and melt kneading them, (2) a method which
5 comprises feeding a part of a block copolymer and a polyphenylene ether from the upstream side feed opening and melt kneading them, and feeding a polyamide and the remainder block copolymer from the downstream side feed opening and melt kneading them, (3) a method which
10 comprises feeding a polyphenylene ether from the upstream side feed opening and melt kneading it, and feeding a block copolymer and a polyamide from the downstream side feed opening and melt kneading them, and the like, and any of these methods may be employed.

15 [0054]

The composition of the present invention obtained in this way can be molded into molded bodies of various parts by using a conventionally known method such as injection molding.

20 Examples of these various parts are electrical equipment parts of motorbikes and automobiles, such as relay block materials, electric and electronic parts such as IC trays, chassis and cabinets of various disk players, OA parts and mechanical parts of various
25 computers and peripheral parts thereof, exterior trim parts such as cowls of motorbikes, bumpers, fenders, door panels, various moldings, emblems, outer door handles, door mirror housings, wheel caps, roof rails

and stay materials therefore, and spoilers of automobiles, interior trim parts such as instrument panels, console boxes and trims, etc.

[0055]

5 The resin composition of the present invention is particularly suitable for relay block materials.

 The embodiments of the present invention will be explained in more detail by the following examples and comparative examples.

10 [0056]

 [Mode for Carrying Out the Invention]

 (Starting materials used)

Component (A): Poly(2,6-dimethyl-1,4-phenylene ether)

 PPE-L

15 Reduced viscosity = 0.42 dl/g

 Molecular weight distribution

(weight-average molecular weight/number-average molecular weight) = 2.1

 PPE-H

20 Reduced viscosity = 0.52 dl/g

 Molecular weight distribution = 2.2

 PPE-W

 Reduced viscosity = 0.50 dl/g

 Molecular weight distribution = 2.9

25 [0057]

 Component (B): Polyamide

 PA-1

 Polyamide 6,6

Relative viscosity = 2.85

Concentration of terminal amino groups = 45
milliequivalent/kg

Concentration of terminal carboxyl group = 75
5 milliequivalent/kg

This polyamide contains 100 ppm of copper
element and 3000 ppm of iodine

PA-2

Polyamide 6,6

10 Relative viscosity = 2.60

Concentration of terminal amino group = 30
milliequivalent/kg

Concentration of terminal carboxyl group = 110
milliequivalent/kg

15 PA-3

Polyamide 6,6

Relative viscosity = 3.23

Concentration of terminal amino group = 52
milliequivalent/kg

20 Concentration of terminal carboxyl group = 60
milliequivalent/kg

This polyamide contains 70 ppm of copper
element and 2400 ppm of iodine.

[0058]

25 PA-MB

Polyamide 6

Relative viscosity = 2.60

Concentration of terminal amino group = 50

milliequivalent/kg

Concentration of terminal carboxyl group = 65

milliequivalent/kg

Master pellet containing 900 ppm of copper
5 element and 18000 ppm of iodine in the polyamide.

[0059]

Component (c): Block copolymer

SEBS-1

Structure: Polystyrene-hydrogenated
10 polybutadiene-polystyrene

Number-average molecular weight = 246,000

Number-average molecular weight of one
polystyrene block = 40,600

Total content of styrene components = 33%

15 Amount of 1,2-vinyl = 33%

Hydrogenation rate of polybutadiene portion =
98% or more

SEBS-2

Structure: Polystyrene-hydrogenated
20 polybutadiene-polystyrene

Number-average molecular weight = 76,000

Number-average molecular weight of one
polystyrene block = 11,000

Total content of styrene components = 29%

25 Content of 1,2-vinyl bond = 32%

Hydrogenation rate of polybutadiene portion =
98% or more

SEBS-3

- Structure: Polystyrene-hydrogenated
polybutadiene-polystyrene
- Number-average molecular weight = 170,000
- Number-average molecular weight of one
- 5 polystyrene block = 29,800
- Total content of styrene components = 35%
- Amount of 1,2-vinyl = 38%
- Hydrogenation rate of polybutadiene portion =
98% or more
- 10 This block copolymer contains 35% by weight of
a paraffinic oil.
- [0060]
- The molecular weight distribution of the
polyphenylene ether and the number-average molecular
- 15 weight of the block copolymer were measured by an
ultraviolet spectrodetector (UV-41 manufactured by Showa
Denko K.K.) using a gel permeation chromatography
measuring apparatus (GPC SYSTEM 21 manufactured by Showa
Denko K.K.) and calculated in terms of standard
- 20 polystyrene. [Solvent: chloroform; temperature: 40°C;
columns: sample side (K-G, K-800RL, K-800R), reference
side (two of K-805L); flow rate: 10 ml/min.; measurement
wavelength: 254 nm; pressure: 15-17 kg/cm²]
- [0061]
- 25 Evaluation tests in the Examples and
Comparative Examples were conducted in the following
manner.
- (1) Spiral flow length (flowing distance in

the mold)

This was evaluated using an injection molding machine (IS-100GN: manufactured by Toshiba Machine Co., Ltd.) with a mold for spiral flow adjusted to a temperature of 80°C setting the cylinder temperature at 280°C. The mold for spiral flow was a mold designed so as to provide a spiral flowing from the central part of the mold and having a width of spiral flow path of 4 mm and a thickness of 3 mm.

10 [0062]

Furthermore, since the spiral flow length is greatly affected by mold temperature, in order to standardize the mold temperature conditions, the molding cycle was fixed to 30 seconds/1 shot in total of 10 seconds of injection time, 15 seconds of cooling time, 1 second of interval and 4 seconds for opening and closing of the mold, and an arithmetical average value of flowing distances in moldings of final 10 shots after carrying out the molding of 50 shots was calculated as the flowing distance in the mold.

[0063]

(2) Post-deformation of the molded product at heating

A relay block shown in FIG. 1 was molded using an injection molding machine (IS-80EPN). (setting of cylinder temperature: 280°C, mold temperature: 80°C, injection time: 15 seconds, cooling time: 20 seconds, injection pressure: 1.1 time the pressure under which no

molding sink was present on the molded piece, injection rate: 60%)

The molded relay block was left to stand in an environment of 23°C and 50%RH for 24 hours, then put in an oven set at 120°C for 30 minutes and taken out, and then left to stand in an environment of 23°C and 50%RH for 24 hours.

[0064]

Thereafter, the size of warpage which occurred at the A-B part in FIG. 1 which was actually to be straight line was measured and was taken as a post-deformation of the molded product at heating.

[0065]

(3) The number of wrinkle patterns

For measuring deformation of the molded piece, the appearance of the molded relay block was observed and formation of wrinkle patterns was examined. When wrinkle patterns were observed, the number of the wrinkle patterns was counted.

[0066]

(4) Tensile strength retention rate after heat aging

A piece for tensile test in accordance with ASTM D-638 was molded using an injection molding machine (IS-80EPN). (setting of cylinder temperature: 280°C, mold temperature 80°C, injection time: 15 seconds, cooling time: 20 seconds, injection pressure: 1.1 time the pressure under which no molding sink was present on

the molded piece)

The resulting molded piece was subjected to heat aging at 150°C for 2000 hours, and then the test piece was taken out and was left to stand in an aluminum moisture proof bag for 48 hours in an atmosphere of room temperature of 23°C. The test piece was subjected to tensile test in accordance with ASTM D-638. The tensile strength retention rate after the heat aging was obtained by the following formula based on a tensile strength of the composition subjected to no heat aging.

[0067]

In order to reduce the error, measurement of tensile strength after heat aging was conducted at 12 points in n number, and the retention rate was calculated from the average of 10 points excluding the maximum value and the minimum value. The retention rate before the heat aging was the average value of 5 points in n number.

Tensile strength retention rate = $\frac{(TS-A)}{(TS-B)} \times 100$ [in the formula, TS-A denotes tensile strength after heat aging and TS-B denotes tensile strength before heat aging.]

[0068]

[Examples 1-4 and Comparative Examples 1-3]

The cylinder temperature of a twin-screw extruder [ZSK-40 manufactured by Werner & Pfleiderer GmbH (Germany)] having one feed opening on the upstream side and one feed opening in the central part of the

extruder was set at 320°C in the area from the upstream side feed opening (hereinafter abbreviated as "main-F") up to the central feed opening (hereinafter abbreviated as "side-F") of the extruder and at 280°C in the area
5 from side-F up to the die.

In accordance with the proportions shown in Table 1, PPE, SEBS and 0.2 part by weight of maleic anhydride (hereinafter abbreviated as "MAH") as a compatibilizer which were uniformly mixed were fed from
10 main-F, and PA in the proportion shown in Table 1 was fed from side-F, and these were melt kneaded to obtain pellets.

[0069]

The screw revolution speed was 300 rpm, and
15 the discharging rate was 80 kg/h. Moreover, residual volatile matter and residual oligomer were removed by carrying out vacuum suction from openings provided at a barrel just before the barrel having the side-F and the barrel just before the die. The degree of vacuum in
20 this case was -700 mmHg.

The pellets obtained under various conditions were used as samples for various measurements.

[0070]

Table 1

Unit	Example 1	Example 2	Example 3	Example 4	Comparative Example 1	Comparative Example 2	Comparative Example 3
PPE-L	30		15	30		25	
PPE-H					30		55
PPE-W		30	15				
SEBS-1	10	10	10		10		
SEBS-2						15	5
SEBS-3				10			
MAH	0.2	0.2	0.2	0.2	0.2	0.2	0.2
PA-1	60					60	40
PA-2		55	55	55			
PA-3					60		
PA-MB		5	5	5			
SEBS/(SEBS+PPE)	0.25	0.25	0.25	0.18	0.25	0.38	0.08
Spiral flow length cm	88	92	90	93	57	92	37
Post-deformation of molded product after heating mm	0.3	0.4	0.4	0.1 or less	1.0	1.3	1.5
The number of wrinkle patterns	4	2	No	No	15	8	Many

Continued

Tensile strength before heat aging MPa	61	62	61	60	60	51	69
Tensile strength after heat aging MPa	49	53	54	55	47	40	28
Tensile strength retention rate %	80	85	89	92	78	78	41

[Effects of the Invention]

The composition of the present invention and the relay block comprising the composition of the present invention can solve simultaneously the problems
5 such as insufficient flowability in the mold, post-deformation of the molded products during heat aging, formation of wrinkle patterns and reduction of tensile strength due to heat aging, as compared with the conventional technologies.

10 [Brief Description of the Drawing]

[FIG. 1]

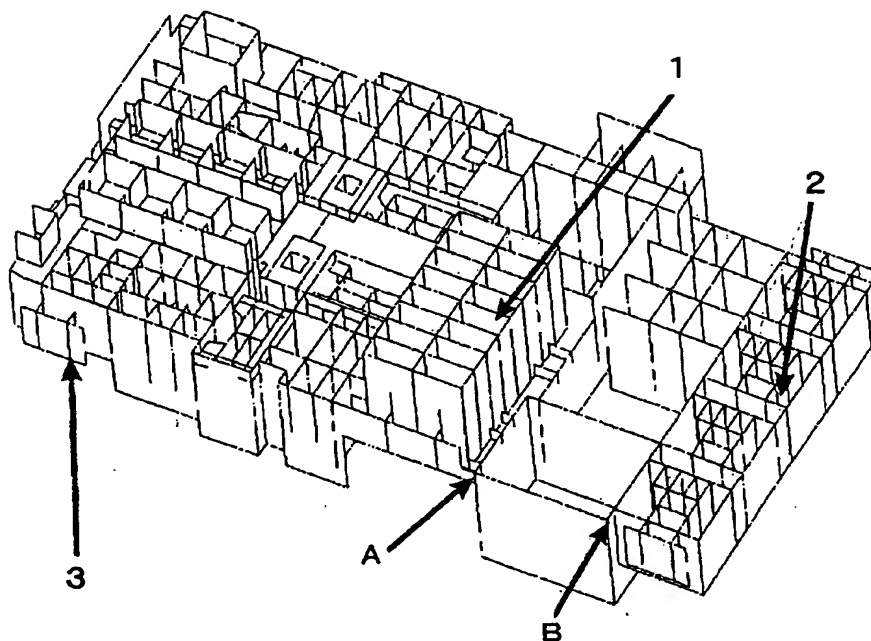
This shows external appearance of the relay block molded in Examples.

[Description of Reference Numerals]

- 15 1: Fuse socket part
2: Relay socket part
3: The part of fitting to car body

Symbols A and B: In measurement of post-deformation of the molded product due to heat aging in
20 Examples, the deformation of the straight line portion comprising A and B was taken as the post-deformation.

【書類名】 図面 Drawing
Kind of Document
【図1】 Document
FIG. 1



[Kind of Document] Abstract

[Abstract]

[Problem] To provide a resin composition highly inhibited from insufficient flowability in the mold, post-deformation after molding, formation of wrinkle patterns and reduction of tensile strength due to heat aging, and a relay block comprising the composition.

[Solution] In a composition comprising a polyamide having a specific viscosity, a polyphenylene ether having a specific viscosity and a block copolymer having a number-average molecular weight of 170,000 or more, the ratio of the block copolymer to the total amount of the block copolymer and the polyphenylene ether is limited to a specific range.

[Selected Drawing] No

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